Two Scales in Asynchronous Ballistic Annihilation

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The kinetics of single-species annihilation, $A+A\to 0$, is investigated in which each particle has a fixed velocity which may be either $\pm v$ with equal probability, and a finite diffusivity. In one dimension, the interplay between convection and diffusion leads to a decay of the density which is proportional to $t^{-3/4}$. At long times, the reactants organize into domains of right- and left-moving particles, with the typical distance between particles in a single domain growing as $t^{3/4}$, and the distance between domains growing as t. The probability that an arbitrary particle reacts with its $n^{\rm th}$ neighbor is found to decay as $n^{-5/2}$ for same-velocity pairs and as $n^{-7/4}$ for $+-$ pairs. These kinetic and spatial exponents and their interrelations are obtained by scaling arguments. Our predictions are in excellent agreement with numerical simulations.

Single-species diffusion-controlled annihilation, $A+A\to 0$, exhibits classical mean-field kinetics when the spatial dimension $d>2$, in which the concentration $c(t)$ decays as t^{-1}, and nonclassical dimension-dependent kinetics for $d\le 2$ with a slower concentration decay, $c(t)\propto t^{-d/2}$ [1-7]. In one dimension, the geometric restriction to nearest-neighbor interactions leads to relatively large departure from the mean-field kinetics, as well as a spatial organization of reactants. In this well-studied case, it is known that $c(t)$ asymptotically decays as $(Dt)^{-1/2}$, independent of the initial concentration. The complementary situation of single-species annihilation where the reactants move ballistically has recently begun to receive attention [8-12]. Perhaps the simplest example is the deterministic $\pm$ annihilation process, where each particle moves at a constant velocity which may be either $+v$ or $-v$ [8,9]. When the densities of the $+v$ and $-v$ particles are equal, $c(t)$ decays as $(c_0/vt)^{1/2}$.

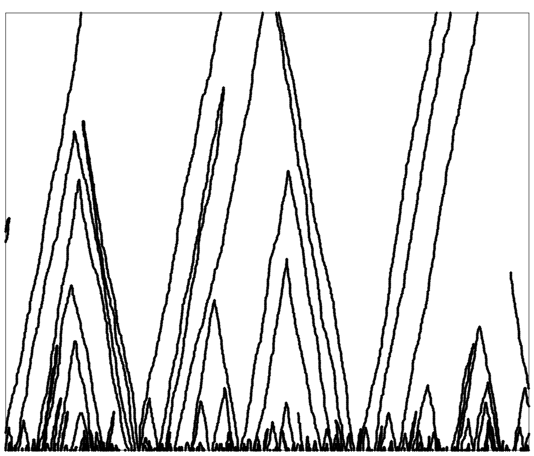

FIG. 1. Space-time evolution of particles in the stochastic $\pm$ annihilation process.

In this letter, we consider single species annihilation when the particle transport is a superposition of convection *and* diffusion — we term this system the stochastic $\pm$ annihilation process (Fig. 1). Although the concentration decays as $t^{-1/2}$ when only one of the transport mechanisms — either convection or diffusion — is operative, the combined transport process leads to a faster concentration decay of $t^{-3/4}$ [10]. Our goal is to understand this unusual decay law and its attendant consequences on the spatial distribution of reactants. While there has been fragmentary mention of some aspects of this system [7,10], here we give primarily new results and a self-contained account of the basic phenomena.

To set the stage for our approaches and results in the stochastic $\pm$ annihilation process, it is first helpful to provide a simple derivation for the decay of $c(t)$ in the deterministic $\pm$ process. Let us consider a system where particles are placed with concentration c_0 in a box of size L, and denote by $c(L,t)$ the time dependent concentration. Initially, there are $N=c_0L$ particles, and the difference between the number of right and left moving particles is of the order of $\Delta N=|N_+-N_-|\sim\sqrt{N}$. Eventually, all particles who belong to the minority-velocity species are annihilated and thus $c(L,t=\infty)\sim\Delta N/L\sim(c_0/L)^{1/2}$. We assume a scaling from for the concentration, $c(L,t)\sim(c_0/L)^{1/2}f(z)$ with $z=L/vt$. According to the above argument, $f(z)\to$ const. in the $z\to 0$ limit. Conversely, in the short time limit, $z\to\infty$, the concentration cannot depend on the box size, so that $f(z)$ must be proportional to $z^{1/2}$. Thus we find

$$c(t)\sim\left(\frac{c_0}{vt}\right)^{1/2}. \tag{1}$$

As a consequence, the system organizes into right- and left-moving domains whose size is of the order of vt.

In the diffusive case, either one particle or no particles survive the annihilation process in a finite box, depending on the parity of the initial number of particles. Following the above line of reasoning, we may write the scaling ansatz $c(L,t)\sim L^{-1}f(z)$ with $z=L/\sqrt{Dt}$. Here the relevant time dependent length scale is $\sqrt{Dt}$. In the limit $z\to 0$, the concentration is independent of L, thereby implying $f(z)\sim z$. Therefore the time dependent concentration is given by

$$c(t) \sim \left(\frac{1}{Dt}\right)^{1/2}. \qquad (2)$$

The crucial new feature in the stochastic $\pm$ annihilation process is that particles with the same velocity can mutually annihilate because of their interaction which is driven by diffusion (Fig. 1). A useful way to determine the decay in this process is to consider separately the role of convection and diffusion on the kinetics. Because of the convection, particles organize into right-moving and left-moving domains as outlined above. Inside each domain, however, diffusive annihilation between same-velocity particles takes place. We assume that the diffusive annihilation mechanism leads to an effective time dependent "initial" concentration, $c_0(t) \sim (Dt)^{-1/2}$, which plays the role of c_0 in Eq. (1). Thus we obtain

$$c(t) \sim \left(\frac{1}{D\, v^2\, t^3}\right)^{1/4}. \qquad (3)$$

Intriguingly, the concentration in the stochastic $\pm$ annihilation process is predicted to decay as $t^{-3/4}$ even though $c(t)$ decays as $t^{-1/2}$ if either diffusion only or convection only is the transport mechanism.

An alternative method to determine the decay law, which provides additional insight into the relative effects of diffusion and convection, is dimensional analysis. If the particle diffusion coefficient is D, then the stochastic $\pm$ process is fully characterized by the initial concentration c_0, the velocity v, and D. From these parameters, the only variable combinations with the dimensions of concentration are c_0, $1/vt$, and $1/\sqrt{Dt}$. On physical grounds, we anticipate that these three concentration scales should enter multiplicatively so that the time-dependent concentration can be expressed in a conventional scaling form. Accordingly, we write the time dependent concentration in the form

$$c(t) \sim (c_0)^\rho \left(\frac{1}{vt}\right)^\sigma \left(\frac{1}{\sqrt{Dt}}\right)^{1-\rho-\sigma}, \qquad (4)$$

in which the dimension of the right-hand side is manifestly a concentration. The exponents ρ and σ can be now determined by requiring that the above expression for $c(t)$ matches with: (a) the diffusion-controlled behavior $c(t) \to (Dt)^{-1/2}$ for $t < \tau_v \simeq D/v^2$, which is the characteristic time below which the drift can be ignored for a particle which undergoes biased diffusion; and (b) the ballistically-controlled behavior $c(t) \to (c_0/vt)^{1/2}$ when $t < \tau_D \simeq 1/(Dc_0^2)$, which is the time for adjacent particles to meet by diffusion. Thus by matching Eq. (4) with $(Dt)^{-1/2}$ at τ_v, one obtains $\rho = 0$, and then matching Eq. (4) with $(c_0/vt)^{1/2}$ at τ_D gives $\sigma = 1/2$. This then reproduces Eq. (3).

To test this decay law, we performed Monte Carlo simulations using the following realization of the reaction process. Initially all sites are occupied with either a + or a − particle with equal probabilities. These signs, which indicate the velocity direction of each particle, remain fixed during the particle lifetime. A simulation step consists of picking a particle at random and moving it a single lattice site in the direction of its velocity. If the target site is occupied, then both particles are removed from the system. Time is updated by the inverse of the number of particles. The simulation was carried up to 10^5 time steps on a periodic chain of 10^6 sites and an average over 10^3 realizations was performed. The data for $c(t)$ is strikingly linear over a substantial time range on a double logarithmic scale (Fig. 2). The local two-point slopes of the data in the time range $10^2 \lesssim t \lesssim 5 \times 10^4$ give an exponent value of 0.745. We interpret the constancy of this data as evidence that the actual value of the exponent is 3/4. It is worth noting that a Padé analysis of the exact short-time power series gives an estimate for the decay exponent of approximately 0.72 [13]. This provides a rough estimate for the magnitude of the variation of the effective exponent between the early time and asymptotic regimes.

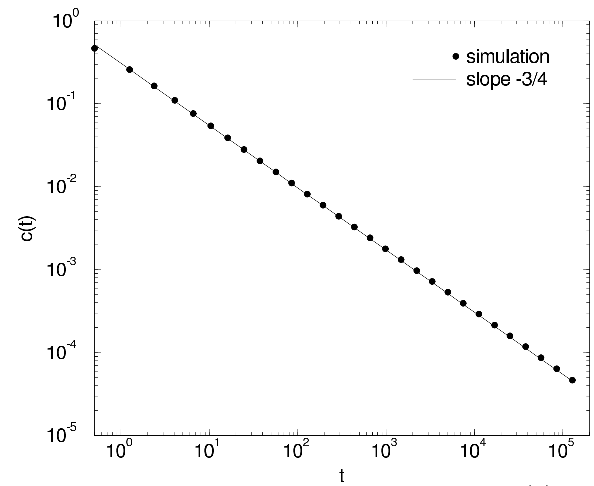

FIG. 2. Simulation data for the concentration (•) versus time on a double logarithmic scale. A line of slope −3/4 is shown for reference.

Having established the decay exponent numerically, it is of interest to consider the consequences of this unusual decay law on the spatial distribution of reactants. In particular, since $c(t)$ decays as $t^{-3/4}$, one might expect that the average separation between nearest-neighbor particles grows as $t^{3/4}$. However, if there remains any vestige of the domain organization that is associated with the deterministic $\pm$ process, then more than one length scale may be needed to characterize this spatial distribution. Such multiscale behavior has been observed previously in diffusive two-species annihilation [14] and the associated consequences lead to new insights about the system. To investigate possible multiscale behavior in the stochastic $\pm$ annihilation process, we introduce the following distance scales (Fig. 3):

$$\langle x_{++}(t)\rangle \sim t^{\nu_{++}}, \quad \langle x_{+-}(t)\rangle \sim t^{\nu_{+-}},$$

$$\langle x_{-+}(t)\rangle \sim t^{\nu_{-+}}, \quad \langle x_{\rm dom}(t)\rangle \sim t^{\nu_{\rm dom}}, \tag{5}$$

which are defined to be, respectively, the average distance between neighboring same-velocity pairs, $+-$ pairs, $-+$ pairs, and the average length of a domain of same velocity particles.

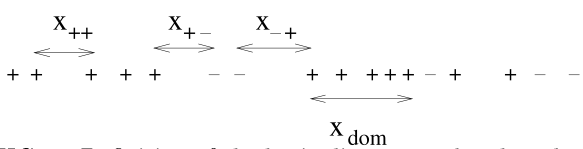

FIG. 3. Definition of the basic distance scales that characterize the spatial organization in the stochastic $\pm$ annihilation process.

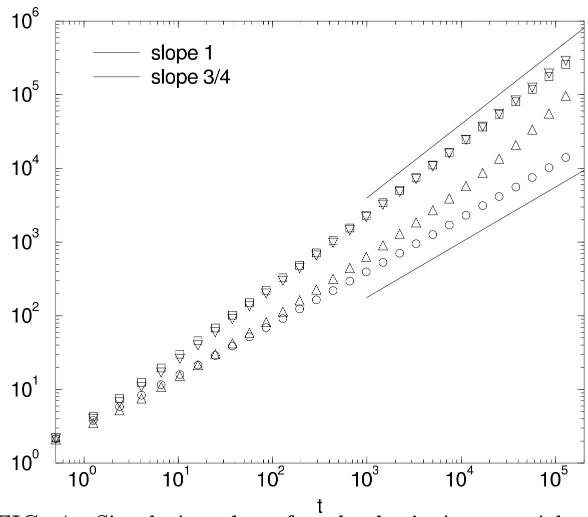

FIG. 4. Simulation data for the basic interparticle distances $\langle x_{++}(t)\rangle \sim t^{\nu_{++}}$ ($\circ$), $\langle x_{+-}(t)\rangle \sim t^{\nu_{+-}}$ ($\triangle$), $\langle x_{-+}(t)\rangle \sim t^{\nu_{-+}}$ ($\square$), and $\langle x_{\rm dom}(t)\rangle \sim t^{\nu_{\rm dom}}$ ($\bigtriangledown$). Lines of slopes 3/4 and 1 are also shown for reference.

Our Monte Carlo data for these length scales exhibits considerable curvature on a double logarithmic scale (Fig. 4). Thus to estimate the asymptotic behavior, we studied the systematic variation of the slopes of linear least-squares fits as the data at the earliest times are progressively eliminated. The effective exponents obtained in this manner vary considerably; for example, for $\langle x_{++}(t)\rangle$, the effective exponent systematically increases, but at a progressively slower rate, from 0.699 to 0.734. Together with relatively strong numerical evidence that the concentration decays as $t^{-3/4}$, we conclude that the actual value of ν_{++} is 3/4. This accords with the expectation that $\langle x_{++}(t)\rangle$ should scale as $1/c(t)$. Similar finite time corrections occur in the exponent estimates for the remaining length scales defined above. For these cases, the effective exponent values are all increasing as short-time data are systematically deleted and it appears that ν_{+-}, ν_{-+}, and $\nu_{\rm dom}$ are all very close to 1, asymptotically. That is, the corresponding lengths are governed by the ballistic particle motion, but again with considerable finite time corrections. The case of $\langle x_{+-}(t)\rangle$ is especially problematic, as the effective exponent changes from approximately 0.80 to 0.93 over the time range covered by our simulation. Evidently, more extensive simulation would be needed to determine the asymptotic exponent values unambiguously by simulation alone.

A new useful way to characterize the spatial range of bimolecular reactions is the collision probability, $P(n)$, defined as the probability that the reaction partner of a given particle is its $n^{\rm th}$ neighbor. Eventually, every particle reacts with some collision partner in one dimension and the distribution of the distances between partners provides a measure of the reaction "efficiency". In the deterministic $\pm$ process, for example, this probability can be obtained analytically [8,9,15]. Let us denote the velocity of the $n^{\rm th}$ neighbor by $v_n = \pm 1$, and the local velocity sum by $S_n = \sum_{i=0}^{n} v_i$. A right moving particle initially at the origin reacts with its $(2n+1)^{th}$-neighbor if: (a) $S_l > 0$ for $l = 0, 1, \ldots, 2n$, and (b) $S_{2n+1} = 0$. This quantity is precisely the same as the first-passage probability for a random walk which starts at the origin to return to the origin for the first time after $2n$ steps. Because of this equivalence to an exactly soluble first-passage problem [16], one has $P(2n) = 0$ and $P(2n+1) = 2^{-2n-1}(2n)!/n!(n+1)!$. In the limit $n \to \infty$, the probability that a given particle collides with its $n^{\rm th}$-neighbor is given by

$$P(n) \propto n^{-3/2}. \tag{6}$$

Motivated by this power law dependence, we assume, in general, that $P(n) \sim n^{-\gamma}$. The exponent γ can be related to other fundamental exponents of reaction processes, namely, the concentration decay exponent α, defined by $c(t) \sim t^{-\alpha}$, and the correlation exponent β, defined by $\xi(t) \sim t^{\beta}$. Here $\xi(t)$ refers to the distance over "information" about the reactants spread. In a time t, only particles within a domain of linear size $\xi(t)$ are eligible to react and thus the surviving fraction, or concentration, is

$$c(t) \sim \int_{\xi(t)} dn\, P(n) \sim \int_{\xi(t)} dn\, n^{-\gamma} \sim t^{\beta(1-\gamma)}. \tag{7}$$

Consequently, we find the exponent relation

$$\gamma = 1 + \alpha/\beta. \tag{8}$$

For the deterministic $\pm$ process, $\alpha = 1/2$ and $\beta = 1$ [8,9], and the exact $\gamma = 3/2$ of Eq. (6) is recovered. As an illustration, consider, for example, single-species diffusion-limited annihilation. The decay and correlation exponents are $\alpha = 1/2$ and $\beta = 1/2$, leading to $\gamma = 2$ from Eq. (8). Preliminary simulations appear to confirm this result. Similarly, for two-species annihilation, α is now equal to 1/4 while β remains 1/2 so that $\gamma = 3/2$.

Let us now consider the behavior of the collision probability in the stochastic $\pm$ annihilation process. In this case, the existence of two length scales in the system suggests that it is necessary to make a distinction between reaction events that involve particles of the same and of different velocities. We therefore define a ballistic correlation scale $\xi_{+-}(t) \sim t^{\beta_{+-}}$, with $\beta_{+-} = 1$, which is associated with $+-$ collisions, *i.e.*, annihilation events between opposite velocity particles. Invoking the scaling relation Eq. (8), we thus find $P_{+-}(l) \sim l^{-\gamma_{+-}}$ with $\gamma_{+-} = 7/4$. Similarly, there is a diffusive length scale $\xi_{++}(t) \sim t^{\beta_{++}}$, with $\beta_{++} = 1/2$, corresponding to annihilation events between same-velocity particles. In this case, Eq. (8) gives $\gamma_{++} = 5/2$. To summarize, we obtain

$$P(n) \sim P_{+-}(n) \sim n^{-3/4} \quad P_{++}(n) = P_{--}(n) \sim n^{-5/2}. \tag{9}$$

This behavior is consistent with our Monte Carlo simulation data (Fig. 5). Notice that over large distances, annihilation between opposite velocity particles dominates, as one would naively expect.

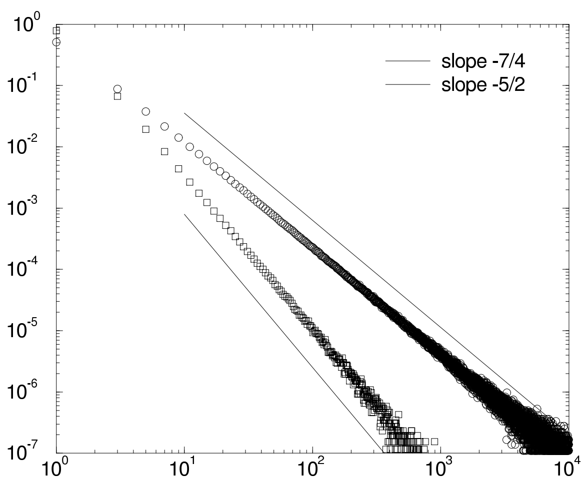

FIG. 5. Simulation data for $P_{+-}(n)$ ($\circ$) and $P_{++}(n)$ ($\square$) on a double logarithmic scale. Lines of slopes $-7/4$ and $-5/2$ are drawn as guides to the eye.

Our results can also be generalized to arbitrary spatial dimension $d > 1$. In this case, it is necessary to ascribe a finite, non-zero radius R to the particles so that there is a finite collision cross section for particles to actually meet. Let us consider the anisotropic system in which particles undergo isotropic Brownian motion with diffusivity D, and a drift along the $\hat{x}$ axis only, with the velocity taking on the value $\pm v\hat{x}$ with equal probability. In the ballistic limit ($D \equiv 0$), the concentration decays as $\sqrt{c_0/R^{d-1}vt}$ (since the process is quasi-one-dimensional, the $t^{-1/2}$ decay of the true one-dimensional system is still obeyed). In contrast, for diffusion-controlled annihilation ($v \equiv 0$), the concentration decays as $(Dt)^{-d/2}$ for $d < 2$, and as $(R^{d-2}Dt)^{-1}$ for $d > 2$ (with logarithmic corrections at the critical dimension $d = 2$) [7]. Repeating the analysis detailed previously for the one-dimensional case in the derivation of Eq. (3) from Eqs. (1) and (2), we find the concentration decay

$$c(t) \sim \begin{cases} (R^{2-2d}\, D^d\, v^2)^{-1/4}\, t^{-(d+2)/4} & d < 2; \\ (R\, D\, v)^{-1/2}\, t^{-1} \left[\ln(Dt/R^2)\right]^{1/2}, & d = 2; \\ (R^{2d-3}\, D\, v)^{-1/2}\, t^{-1}, & d > 2. \end{cases} \tag{10}$$

The combined diffusion and ballistic transport does not change the mean-field nature of the annihilation kinetics when $d > 2$ and the classical t^{-1} decay is recovered. For sufficiently low spatial dimension, however, the non-classical behavior arises in which the decay exponent $\alpha = (d+2)/4$. Thus in low spatial dimensions, the interplay between convection and diffusion provides more effective mixing than diffusion or drift alone, and leads to a larger decay exponent than $\alpha_{\rm diff} = d/2$ and $\alpha_{\rm ball} = 1/2$ which arise when only one transport mechanism is operative.

In summary, the stochastic $\pm$ single-species annihilation process exhibits a $t^{-3/4}$ decay of the concentration. This is faster than the $t^{-1/2}$ decay that arises when only one of the constituent transport processes in the stochastic $\pm$ process, either diffusion and deterministic $\pm$ convection, is present. A microscopic understanding of this decay law is lacking, and it seems that a technique beyond those typically used to solve one-dimensional reactive systems would be needed for the stochastic $\pm$ annihilation process. At long times, the system exhibits a spatial organization in which diffusion controls the short distance behavior and convection controls the large distance behavior. We have also introduced the concept of the collision probability, $P(n)$, the probability that a given particle is annihilated by its $n^{\rm th}$-neighbor. For the stochastic $\pm$ process, this probability is further discriminated by annihilation by same-velocity and opposite velocity pairs. These two probabilities decay as $P_{++}(n) \sim n^{-5/2}$ and $P_{+-}(n) \sim n^{-7/4}$, respectively. It will be interesting to study the collision probability in other reaction processes such as diffusive driven single-species annihilation.

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